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Ferrite-Ferroelectric Composite Materials for Microwave and Millimeter Wave Device Applications

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A series of composite ferroelectric ferrite materials has been fabricated in order to investigate the basic properties which may lead to new microwave capabilities. The ferrite component was a Trans-Tech TT2-111 NiZn ferrite obtained in powder form. The ferroelectric component consisted of Paratek Type A and Type B Parascan™ materials. The ferroelectric component was nominally barium strontium titanate. The composite materials were prepared with 0.3 - 50 wt% ferrite, along with a 100 wt% ferrite material fired from the TT2-111 powder. These materials were used for low frequency and microwave dielectric constant and loss measurements, static magnetic characterization, ferromagnetic resonance, and high field microwave susceptibility measurements. The measurements indicate that such composites can, in fact, be fabricated and that the basic properties can be tuned through the mixing. Although much more materials development remains, the samples demonstrate that one can obtain a field tunable magnetic response through the ferrite component and a variable dielectric constant through the mixing. The specific microwave data show that the static magnetic properties scale with the ferrite content for loadings above 10 wt% or so. The microwave response data appear to scale with the ferrite loading, except for one critical result at 25 wt% loading. Further work is needed to determine the extent of a possible response enhancement for the ferrite based on the field concentrations induced by the ferroelectric. Further work is also needed to optimize the processing parameters and specific materials choices to obtain low dielectric loss and narrow ferromagnetic resonance linewidths.

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ABSTRACT

A series of composite ferroelectric ferrite materials has been fabricated in order to investigate the basic properties which may lead to new microwave capabilities. The ferrite component was a Trans-Tech TT2-111 NiZn ferrite obtained in powder form. The ferroelectric component consisted of Paratek Type A and Type B Parascan™ materials. The ferroelectric component was nominally barium strontium titanate. The composite materials were prepared with 0.3 - 50 wt% ferrite, along with a 100 wt% ferrite material fired from the TT2-111 powder. These materials were used for low frequency and microwave dielectric constant and loss measurements, static magnetic characterization, ferromagnetic resonance, and high field microwave susceptibility measurements. The measurements indicate that such composites can, in fact, be fabricated and that the basic properties can be tuned through the mixing. Although much more materials development remains, the samples demonstrate that one can obtain a field tunable magnetic response through the ferrite component and a variable dielectric constant through the mixing. The specific microwave data show that the static magnetic properties scale with the ferrite content for loadings above 10 wt% or so. The microwave response data appear to scale with the ferrite loading, except for one critical result at 25 wt% loading. Further work is needed to determine the extent of a possible response enhancement for the ferrite based on the field concentrations induced by the ferroelectric. Further work is also needed to optimize the processing parameters and specific materials choices to obtain low dielectric loss and narrow ferromagnetic resonance linewidths.

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I. RESEARCH RESULTS

A. Problem Statement and Overview

This short term innovative research (STIR) final report summarizes results from a collaborative effort between Colorado State University, Department of Physics, and PARATEK Inc. The main objective was to investigate the dielectric and magnetic properties of ferrite/ferroelectric composite materials for microwave and millimeter wave device applications. The investigation is motivated by the promising possibility to combine the large phase shift and power limiting properties of ferrite materials with the voltage tunable properties of the ferroelectric component. The lower dielectric constant of the ferrite could also serve to provide a better impedance match for the composite than for the ferroelectric alone. The combination would yield an optimum material for microwave signal processing applications.

The main magnetic scenario considered in this initial STIR program was that the microwave electric field concentrations induced by the higher dielectric constant of the ferroelectric would lead to an enhancement of the microwave magnetic field in the material. This enhancement would lead to a larger magnetic response and a larger effective susceptibility. The strategy was to (i) prepare a range of composite materials with different ferrite loadings, (ii) measure static magnetic properties and the microwave response, (iii) examine the static response to determine if these properties scale with the loading, and (iv) examine the microwave response to see if these properties either scaled with the loading or showed a larger effect. A larger microwave response than predicted from scaling would indicate the ferroelectric enhancement proposed above.

The specific microwave data show that the static magnetic properties scale with the ferrite content for loadings above 10 wt% or so. The microwave response data appear to scale with the ferrite loading, except for one critical result at 25 wt% loading. Further work is needed to determine the extent of a possible response enhancement for the ferrite based on the field concentrations induced by the ferroelectric. Further work is also needed to optimize the processing parameters and specific materials choices to obtain low dielectric loss and narrow ferromagnetic resonance linewidths.

B. Summary of Results

1. Materials

The composite materials were prepared by Drs. Somnath Sengupta and Louise C. Sengupta at Paratek, Inc. The samples for this program consisted of thick disks of Type A and Type B Parascan^{TM} tunable dielectric materials, nominally ferroelectric barium strontium titanate, with different loadings of NiZn ferrite. The ferrite was a standard commercial Trans-Tech TT2-111 material in powder form.

Different weight percentages of the TT2-111 ferrite powder (0.3 wt%, 1.0 wt%, 5.0 wt%, 10 wt%, 25 wt%, and 50 wt%) were mixed with the tunable dielectric materials. The mixtures were ball-milled for 24 hours in a slurry of ethanol with an alumina grinding media. After drying, the

powder mixtures were sieved. The specimens were then pressed and subsequently sintered. The densities of specimens sintered at different temperature were determined from the measured weights and volumes of the samples. The sintering temperatures were then optimized to obtain the highest density. Fired samples of the pure TT2-111 powder without any dielectric component were prepared in a similar manner. The fired samples were all in the form of approximately one inch diameter, one quarter inch thick disks.

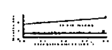
Type A composites were used for the dielectric measurements performed at Paratek. The samples which were used for the low frequency dielectric measurements were screen printed with Ferro #T2026 ink and fired at 760°C. The 10 GHz dielectric measurements were made without electrodes.

Most of the magnetic measurements were made at Colorado State University on both Type A and Type B composites. Some magnetization data were also obtained at the National Institute of Standards and Technology in Boulder, Colorado with the assistance of Dr. Ronald B. Goldfarb. For these measurements, small cubes or spheres were fabricated from the fired disks. The Type A samples were generally harder and it was more difficult to cut cubes or grind spheres. Some of these samples also appeared to have excessive microwave loss. All of the magnetic measurements discussed below were made on Type B materials. The static measurements were made by vibrating sample magnetometry. The microwave measurements were made by standard cavity or shorted wave guide techniques.

Table I summarizes some basic properties of the ferrite component. More detailed results on dielectric, static magnetic, and microwave magnetic properties will be discussed shortly. The main point of the table is to establish the basic saturation induction $4\pi M_s$ and density for the

Table I. Selected properties of TT2-111 ferrite material.

Sample	Saturation Induction $4\pi M_s$ (kG)	Specific Magnetization σ (emu/g)	Density ρ (g/cm^3)	10 GHz FMR Linewidth(1) (Oe)
Paratek fired TT2-111 powder	4.32(2) 4.33(3)	76.9(4)	4.48(5)	1200(6)
Trans-Tech fired TT2-111	4.74(3)	76.5(7)	4.90(8)	
Trans-Tech specification sheet	5.03		3.94	154

(1) Ferromagnetic resonance (FMR) half power linewidth.
(2) Measurement made on sphere. Based on magnetization vs. field response and a sphere demagnetizing factor of 1/3.
(3) Based on indicated specific magnetization σ, density ρ, and the connection  .
(4) Measured on a sphere in a 10 kOe external magnetic field.
(5) Measured on Paratek fired disk.
(6) Shorted wave guide measurement at 10 GHz.
(7) Measured on a 0.1 cm^3 cube in a 5 kOe external field.
(8) Measured on a 30 cm^3 block.

ferrite. These parameters will be needed for the high field microwave response analysis. As indicated above, Trans-Tech TT2-111 is a NiZn ferrite. The Zn content gives the relatively large saturation induction of 4.3 kG. The measured density of the Trans-Tech fired material is somewhat higher than that for the Paratek fired material. This indicates a porosity of about 2-3% for the Paratek fired material. The large ferromagnetic resonance (FMR) linewidth in the Paratek material is consistent with this indication. Where needed for the analysis, the Paratek $4\pi M_s$ value of 4.33 kG and density value of 4.48 g/cm^3 will be used. Note that the $4\pi M_s$ value obtained from the specific magnetization and density measurements match the value obtained from the initial slope of the magnetization vs. field response and the saturation field. This means that the sample response to the field is in agreement with simple micromagnetic and demagnetizing field considerations.

Figure 1 shows a measured hysteresis loop for a sphere of the Paratek base TT2-111 material. This material will be referred to as the sample with "100 wt% loading." The figure shows the magnetic induction $4\pi M$ as a function of the applied external magnetic field. These $4\pi M$ values were derived from the calibrated magnetic moment vs. field data and the sample volume as determined from the sphere diameter. As indicated above, the saturation induction of 4.3 kG is somewhat smaller than the $4\pi M_s$ for the Trans-Tech fired TT2-111 material. The "knee" of the hysteresis loop occurs at an external field of approximately $4\pi M_s/3$, so that the saturation occurs in the correct field range for a sphere sample.

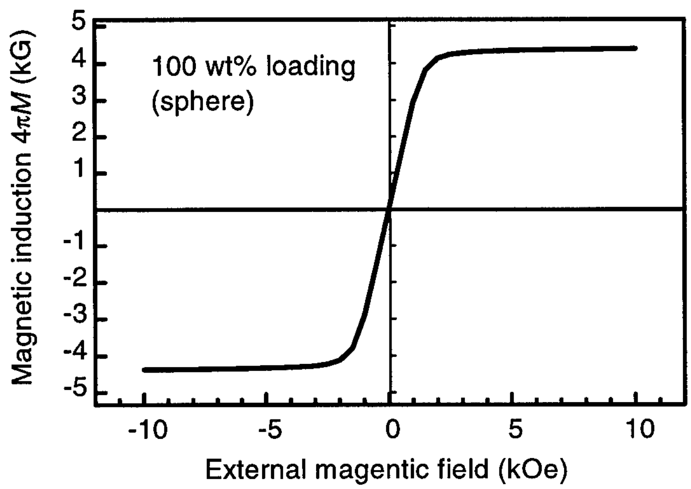


Fig. 1. Hysteresis loop of magnetic induction $4\pi M$ as a function of the external magnetic field for a sphere sample of the 100 wt% loading Paratek fired Trans-Tech TT2-111 NiZn ferrite powder.

The starting materials for the magnetic measurements were the one inch diameter , 0.25 inch thick disks from Paratek, fabricated as described above. These disks were cut into small cubes. Some of the cubes were also used to make spheres. Table II shows some parameters for the Type B samples. There were no significant differences in the saturation moments measured on cubes and spheres. Except for the 100 wt% loading sample, most of the measurements were made on cubes.

Table II : Table of Type B samples and nominal specifications

Sample	Loading(1) wt%	Density(1) g/cc	Shape	Mass g	Volume mm^3	Density(3) g/cc	Moment (4) emu	emu/g	$4\pi M_s$ (5) G
B0	0.0	5.25	Block	5.013	938.8	5.32	0	0	0
B1	0.3	4.46	Cube	0.062	13.7	4.53	0.0002	0.003	0.2
B2	1.0	4.45	Cube	0.066	14.6	4.49	0.006	0.09	5
B3	5.0	4.20	Cube	0.044	9.60	4.60	0.044	1.0	58
B4	10.0	4.50	Cube	0.055	13.30	4.16	0.15	2.7	140
B5	25.0	4.59	Cube	0.026	5.60	4.59	0.50	19.4	1117
B6	50.0	-	Cube	0.030	6.60	4.62	1.11	36.3	2107
Ferrite	100.0	4.48 (2)	Sphere	0.113	24.8	4.55	8.68	76.9	4397

(1) Nominal values from Paratek.
(2) Same as in Table I, obtained from magnetization data.
(3) Estimated from the ratio of mass to volume for the indicated sample.
(4) Saturation value obtained for a 10 kOe applied field.
(5) Based on indicated moment in emu and indicated volume.

2. Dielectric properties

The 1 MHz dielectric constant, tunability (%), and dielectric loss measurements were done with a Hewlett Packard (HP) impedance gain/phase analyzer (HP4194A) meter. The 10 GHz measurements were obtained with a 10 GHz cavity resonator and a HP 8722D vector network analyzer. The resonator measurement method gives an accuracy for the dielectric constant determinations in the ± 0.5% range and also provides the sensitivity which is needed for low level dielectric loss tangent measurements above 10^{-5}.

Table III summarizes the dielectric property measurements performed at Paratek. The properties of the A2 type of Parascan™ dielectric show less change with addition of the TT2-111 ferrite. The dielectric constant at 10 GHz changes from 412 for the pure Parascan™ dielectric to about 200 with the incorporation of 10 wt% of TT2-111. The dielectric loss tangent changes from 0.0235 for pure Parascan™ dielectric to about 0.0279 for the 10 wt% TT2-111 composite. As discussed below, one also finds an increase in the electrical loss with increasing ferroelectric component from the microwave cavity measurements. These effects indicate that some optimization of the preparation conditions for the composites is needed. The Trans-Tech specification on the loss tangent for the ferrite component is below 0.001. It should be possible, therefore, to make composites which show no degradation in loss tangent relative to the pure ferroelectric component. The data in Table III, as well as the magnetic data given below, are for one class of Paratek tunable ferroelectric materials and one type of ferrite. Other ParascanTM materials and other ferrites may well yield better properties for tunable microwave device applications.

Table III. Summary of dielectric property measurements

Material composition	Relative dielectric constant at 1MHz	Relative dielectric constant at 10GHz	Dielectric loss tangent (tanδ) at 1MHz	Dielectric loss tangent (tanδ) at 10GHz	Tunability (%)	
					2V/μm	4V/μm
Ferrite	12.6		0.0390			
Type A1	4756		0.0050		54.0%	71.0%
A1+0.3 wt% Ferrite	2748		0.0013		27.0%	49.0%
A1+1.0 wt% Ferrite	1746		0.0012		23.0%	41.0%
A1+5.0 wt% Ferrite	1328		0.0011		7.9%	20.0%
A1+10 wt% Ferrite	1197		0.0019		9.9%	23.0%
A1+25 wt% Ferrite	883		0.0093		13.8%	27.0%
Type A2	395	412	0.0010	0.0235	17.0%	29.0%
A2+0.3 wt% Ferrite	383		0.0006		12.0%	25.0%
A2+1.0 wt% Ferrite	365	342	0.0009	0.0191	11.0%	24.0%
A2+5.0 wt% Ferrite	262	255	0.0018	0.0245	8.0%	19.0%
A2+10 wt% Ferrite	246	203	0.0019	0.0279	9.0%	19.0%
A2+25 wt% Ferrite	154	128	0.0025	0.0301	8.5%	16.0%
A2+50 wt% Ferrite	69		0.0069		3.2%	6.8%

3. Static magnetization as a function of loading

One objective of the static magnetic measurements was to demonstrate that the ferrite powder could be loaded in the ferroelectric matrix, fired appropriately, and still preserve the magnetic moment of the original material. Figure 2 shows specific magnetization as a function of the ferrite loading, based on the data in Table II. The solid line shows a best fit to the data with the added constraint of zero specific magnetization at zero loading. These data show that the specific magnetization scales with the loading, except for the loading values below 10 wt% or so. This indicates that, on the whole, the ferrite was not modified by the processing used to make the composites. For the small loading samples, the magnetizations fall below the linear curve. For these samples, the hysteresis loop response was also weak and distorted, and did not show the magnetic hysteresis character of Fig. 1. These data indicate that for small loadings there is some modification of the ferrite phase which degrades the magnetic response. Further work is needed to define the magnetic response at low loadings.

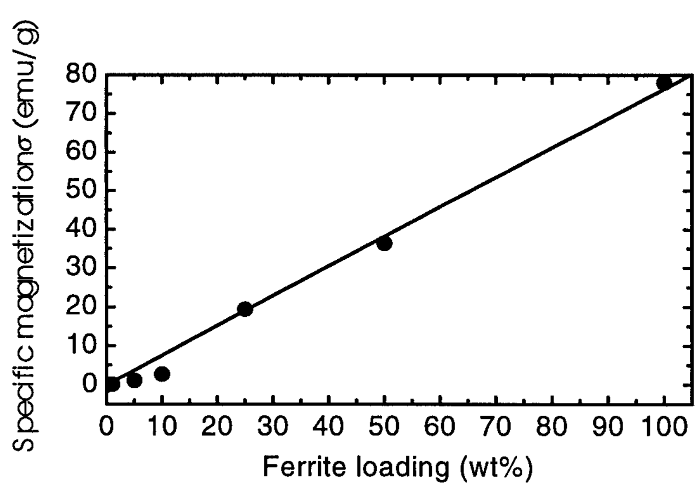


Fig. 2. Specific magnetization as a function of ferrite loading.

Some additional information may be obtained from the hysteresis loop data for intermediate loadings. Data for the composites with 5, 10, 25, and 50 wt% loadings are shown in Fig. 3, along with reference data for 100 wt% loading. These data show that the field position for the knee of the hysteresis loop also scales with the loading. For the 100 wt% loading sample, the knee of the loop is in the 1.5 - 2 kOe range, and this corresponds to the saturation field value of $4\pi M_s/3$ expected for the spherical sample. The problem lies with the lower loadings. The knee of the hysteresis loop for the 25 wt% loading sample, for example, is about 0.3 - 0.5 kOe. At loadings below 25 wt%, the initial slopes increase somewhat with loading. Nevertheless, all of the curves in Fig. 3 have a more-or-less common low field response. As the samples approach saturation, the individual curves simply move off the common low field load line. This gives a saturation field, which *decreases* as the loading decreases. Non-interacting spherical ferrite particles in a nonmagnetic matrix should have a saturation field of $4\pi M_s/3$ which is independent of the loading.

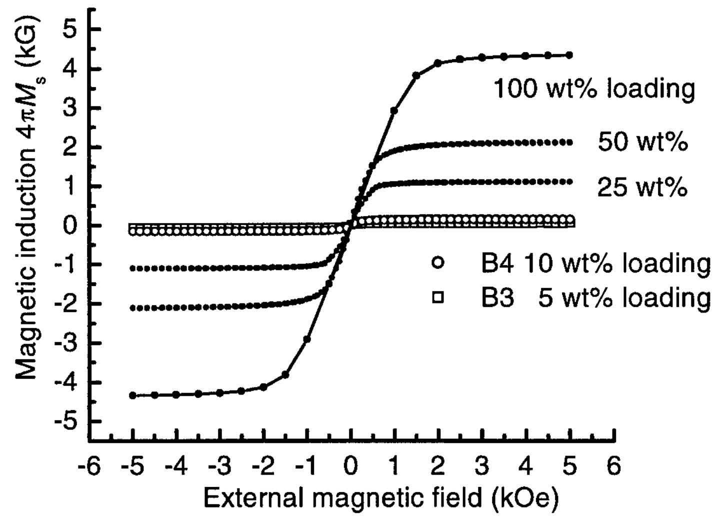


Fig. 3. Saturation induction vs. static external field for the indicated loadings.

The fact that the saturation field scales with loading indicates a microstructure which is more complicated than a simple ferroelectric matrix with spherical non-interacting ferrite particles distributed within the matrix. However, interactions between magnetic grains, considered alone, cannot explain the data either. While interactions could cause a reduced saturation field, as found here, the strength of these interactions would decrease rather than increase for the lower loadings. The data show that the saturation field decreases as the loading decreases.

4. Ferromagnetic resonance response and linewidth

As indicated in Table I, the FMR linewidth of the 100 wt% loading material is well above the expected linewidth for TT2-111 ferrite materials. The apparent porosity from the lower density and lower saturation induction are the most likely sources of this larger linewidth. Figure 4 shows FMR absorption curves for the 10 wt%, 25 wt%, and 50 wt% composite samples, along with a reference FMR curve for the 100 wt% loading sample. The operating point frequency was 10 GHz. The absorption profiles were obtained with a shorted wave guide technique. The FMR absorption profiles consist of the negative going deviations from the base line in each case. One obtains a decrease rather than an increase in the reflected power at FMR because more power is absorbed by the sample.

The main point of the data in Fig. 4 is to demonstrate that the microwave response of the magnetic phase of the composite materials does not correspond to the response expected from simple ferromagnetic resonance considerations. In particular, the FMR response of the 10 wt% and 25 wt% are highly asymmetric, with the FMR peaks significantly shifted from the expected 3.4 kOe values. The fact that there is an FMR response at all indicates that the ferrite component does in fact enter the composite in some simple fashion. The large linewidths and severe distortions in the absorption profiles are indications of additional loss processes due to the composite formation process and a complicated ferrite microstructure. Recall that the static hysteresis response also indicated a complicated ferrite microstructure.

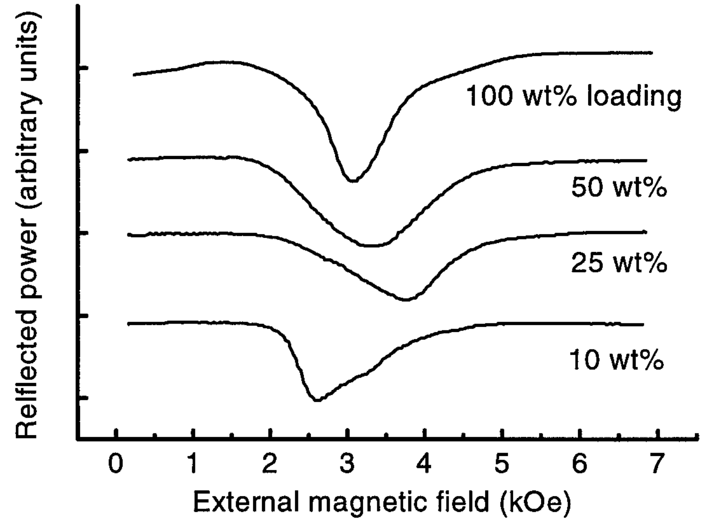


Fig. 4. Ferromagnetic resonance absorption profiles for the three composites with the highest loadings and the 100 wt% loading sample at 10 GHz.

These complications could involve ionic modifications or impurities, different size and shape distributions for the ferrite particles in the ferroelectric matrix, interactions between the ferrite particles, or some sort of magnetic porosity. Based on this preliminary study, it is possible to do no more than speculate on the origin of the responses shown in Fig. 4. One point is clear. The widths of the responses are much larger than the expected 150 - 200 Oe for a dense conventionally fired TT2-111 material and these linewidths show no clear trend as a function of loading. This applies to the 100 wt% loading sample as well as to the composites.

5. The high field microwave response as a function of loading - theory

The FMR data in Fig. 4 show that the effects of microstructure, porosity, and other undetermined effects dominate the resonance response. One way to examine the basic microwave response of an inhomogeneous magnetic system is to measure the high field tails of the FMR absorption and dispersion [Patton, 1975; Truedson *et al.*, 1992, 1993, 1994]. If the distortions to the FMR response in the near resonance field range are viewed in terms of two magnon scattering or inhomogeneous line broadening, one can argue that the high field response corresponds to the undistorted tail of the Lorentzian response associated with some "intrinsic" microwave susceptibility. While this assumption may appear as questionable for the severe FMR profile distortions and large linewidths in Fig. 4, previous work on high field effective linewidths in porous polycrystalline ferrites with highly broadened and distorted lines (Patton, 1975) shows that the high field tail response is still very Lorentzian in character. The high field FMR tail analysis, therefore, can still provide a starting point for an understanding of the effects of loading on the basic microwave response of the composite. As shown below, the dispersion

tail analysis can give an indication of scaling for the composite response. The absorption tail analysis yields a magnetic loss parameter for comparison with linewidths for the base ferrite.

The high field microwave response of the composites with 10 wt%, 25 wt%, and 50 wt% loadings, and that of the base 100 wt% loading material, was evaluated by standard microwave techniques. The evaluations were done at fields which were typically above 5 kOe or so. As just indicated, these high field data avoid, in principle, most of the complications introduced by the large inhomogeneously broadened ferromagnetic resonance (FMR) linewidths and convoluted line shapes shown above. The high field response maps the tail of the FMR response only. The change in the cavity frequency with field maps the real part of the microwave susceptibility or the dispersion. The change in the quality factor, or Q, or the cavity with field yields the tail of the microwave absorption. This section will establish simple working equations for the tail analysis. The details of the derivations and the background physics are discussed in the references cited above. The experimental results are given in the next section.

The connections between the cavity frequency and Q changes with field and the complex microwave susceptibility of the sample under test can be easily obtained from microwave perturbation theory. These connections may be written in terms of two equations.

$$\frac{f - f_N}{f} = -K\, 4\pi\chi' . \tag{1}$$

$$\frac{1}{Q} - \frac{1}{Q_N} = +2\, K\, 4\pi\chi'' . \tag{2}$$

In the above, f_N and Q_N denote the cavity frequency and Q extrapolated to or measured at very high fields, K is the response factor for the sample and the cavity, and $\chi = \chi' - i\chi''$ specifies the complex microwave susceptibility tensor diagonal component for the applicable microwave field direction in the sample. The K parameter depends on the details of the cavity and the active magnetic volume of the sample. For the present purposes, it is useful to write K in the form

$$K = C \frac{m}{\rho_F} L , \tag{3}$$

where C is a cavity parameter, m is the mass of the sample, ρ_F is the density of the ferrite component, and L is the ferrite loading weight fraction. For given sample, the K parameter can be obtained empirically from frequency vs. field data according to the equations to be considered below. One may also obtain K from Eq. (3), with the cavity parameter C obtained from microwave perturbation theory. The C parameter may be written as

$$C = \frac{1}{4\, V_{cav}\, g} \quad , \tag{4}$$

where V_{cav} is the volume of the cavity and g is a factor which relates the cavity energy to the microwave field amplitude. This factor contains only the cavity dimensions and mode parameters. From Eq. (3), one obtains the simple condition, $K/mL = C/\rho_F$. For the cylindrical TE_{011} cavity used here, C is equal to 0.109 cm^{-3}. If one takes the density of the ferrite phase within the dielectric matrix to be the same as that of the 100 %wt ferrite material, that is $\rho_F = 4.48\ \mathrm{g/cm^3}$ as in Table I, then one obtains a theoretical value of 0.0244 for K/mL. This result will be used in the next section to obtain a theoretical response curve for comparison with data, based on the analysis below.

The real part of the susceptibility, χ', corresponds to dispersion. For a spherical shaped isotropic ferrite sample or particle at magnetic fields well above the FMR field, $4\pi\chi'$ takes the simple form

$$4\pi\chi' = \frac{4\pi M_s H}{H^2 - (f/\gamma_{\mathrm{GHz}})^2}. \tag{5}$$

In the above, H is the external applied magnetic field, f is the excitation frequency, and γ_{GHz} is a gyromagnetic parameter which is approximately equal to 2.8 GHz per kOe for most ferrites. Equations (1), (3), and (5) may be combined to yield an operational equation for the cavity frequency vs. field in the form

$$f = f_N - K\, X_F(H,f) = f_N - C\frac{m}{\rho_F}L\ X_F(H,f), \tag{6}$$

where the frequency vs. field scaling function $X_F(H,f)$ is given by

$$X_F(H,f) = \frac{4\pi M_s H f}{H^2 - (f/\gamma_{\mathrm{GHz}})^2}. \tag{7}$$

This $X_F(H,f)$ variable will be termed the dispersion parameter. Equations (6) and (7) provide the basis for the scaling analysis of the microwave data. The ferrite density ρ_F, the magnetization M_s and the gyromagnetic parameter γ_{GHz} are assumed known. All of the other parameters are accessible from experiment. Note that ρ_F and M_s are the density and magnetization, respectively, for the ferrite phase and not the average density and magnetization of the sample as a whole.

It is important to keep in mind that the main objective of these preliminary microwave evaluations was to examine the scaling of the response. One simple way to test this scaling is compute $X_F(H,f)$ vs. field and frequency, and then plot the measured cavity frequency f as a function of X_F. In the high field tail regime, and if the uniform mode FMR dispersion tail response specified by Eq. (5) is valid, f vs. X_F will be a linear function and the slope of the response will correspond to $-K$. The reduced slope parameter K/m will then correspond to CL/ρ_F. If the scaling hypothesis is correct, the data should give a K/m parameter which scales linearly with the loading fraction L for the composite and extrapolates to zero at $L = 0$.

Furthermore, this empirical K/m vs. L plot may be compared directly with the result from perturbation theory, based on the condition established above, namely, $K/m = 0.0244\,L$.

From the above, it is clear that plots of the cavity frequency f vs. the dispersion parameter X_F provide several useful tests. First, a linear response provides proof that (1) the magnetic response actually corresponds to the ferromagnetic dispersion tail and (2) the FMR response is in accord with the assumed uniform mode response. Note that this response will be different for different sample shapes. The above working equations are specifically for spheres. Given a linear response, the slope parameter K provides an automatic empirical magnetic calibration for the cavity. This calibration may be used directly to obtain loss parameters, such as the effective linewidth discussed below, from the data on cavity Q vs. field. This calibration may also be compared with the corresponding value obtained from microwave perturbation theory and Eqs. (3) and (4), as a check on the general validity of the analysis method.

The above scenario is based on six assumptions. (i) Each ferrite particle has a magnetization which is equal to the bulk ferrite value M_s. (ii) The particles are non-interacting. (iii) Each ferrite particle in the matrix is a sphere. (iv) The particles were excited homogeneously by the applied microwave field. (v) The uniform mode ferromagnetic resonance response model based on the torque equation of motion with damping is applicable. (vi) The cavity response can be modeled in the form given by Eqs. (1) and (2), as prescribed by microwave perturbation theory.

For a known ferrite material, there is usually a second step in the analysis which may be used to determine the so called "effective linewidth ΔH_{eff} from the high field tail response for the FMR absorption. The absorption is described by the $4\pi\chi''$ response. For a spherical non-interacting ferrite particle, one may write the high field tail response for $4\pi\chi''$ in the form

$$4\pi\chi'' = \frac{1}{2}\,\frac{4\pi M_s \Delta H_{\mathrm{eff}}\left[H^2 + (f/\gamma_{\mathrm{GHz}})^2\right]}{\left[H^2 - (f/\gamma_{\mathrm{GHz}})^2\right]^2}. \tag{8}$$

Equations (2) and (8) may then be combined to obtain a Q response in the form

$$\frac{1}{Q} = \frac{1}{Q_N} + K\,\Delta H_{\mathrm{eff}}\,X_{\mathrm{Q}}(H,f) = \frac{1}{Q_N} + C\,\frac{m}{\rho_F}\,L\;\Delta H_{\mathrm{eff}}\,X_{\mathrm{Q}}(H,f), \tag{9}$$

where the $1/Q$ scaling function $X_{\mathrm{Q}}(H,f)$ is given by

$$X_Q(H,f) = \frac{4\pi M_s\left[H^2 + (f/\gamma_{\mathrm{GHz}})^2\right]}{\left[H^2 - (f/\gamma_{\mathrm{GHz}})^2\right]^2}\quad. \tag{10}$$

The above equations allow one to combine data on cavity Q vs. field with the cavity frequency vs. field data and determine the high field effective linewidth from the combined tail responses for the absorption and dispersion. For a given sample, the slope from the plot of cavity frequency f vs the frequency scaling function $X_F(H,f)$ gives the calibration parameter

K. Similarly, the slope of the plot of $1/Q$ vs. the $1/Q$ scaling function $X_Q(H,f)$ gives the $K\,\Delta H_{eff}$ product. The ratio of these two slopes then gives the effective linewidth. The significance of this high field effective linewidth is discussed by Patton (1975). In the context of composite materials and the problematic FMR absorption curves noted above, the meaning of ΔH_{eff} will require some additional discussion. These points will be considered in the next section.

It is important to emphasize that the above working equations apply to spherical non-interacting ferrite particles for which the uniform mode FMR response assumptions are valid. If the ferrite particles can not be approximated in this way, the analysis becomes much more complicated. The successes and failures of this simple model will be evident in the next section. The data show that (i) the spherical particle high field effective linewidth analysis provides a reasonable starting point for this study of microwave ferrite ferroelectric composites and (ii) a more sophisticated approach will be needed in order to obtain a quantitative understanding of these materials.

6. The high field microwave response as a function of loading - experiment

Figure 5 shows the results of the tail analysis for the high field dispersion response. The X_F parameter was evaluated from the frequency vs. field data, a saturation induction $4\pi M_s$ value of 4.33 kG, and a gyromagnetic ratio, γ_{GHz}, of 2.8 MHz/Oe. As expected from the response equations given above, the frequency shift, relative to the cavity frequency in the high field limit and normalized to the sample mass, is a linear function of the dispersion tail parameter X_F for all the samples. From Eq. (3), one would expect the slopes of the response, given by K/m, to scale with the loading L. The data show that this is generally the case, except that the responses for the 25 wt% and 50 wt% loading samples are not in the expected order.

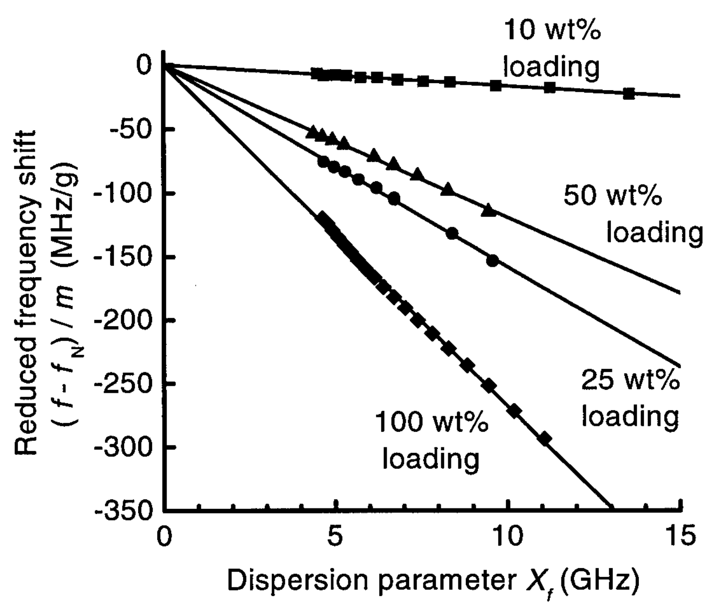


Fig. 5. Reduced data from the tail analysis for the real part of the microwave susceptibility vs. field for the 10 wt%, 25 wt%, 50 wt%, and 100 wt% loadings.

Figure 6 shows the slopes of the linear responses in Fig. 5 as a function of loading. The solid line shows the theoretical response. This line corresponds to the $K/m = 0.0244\,L$ response obtained in the previous section. Except for the 25 wt% sample, the data show that K/m scales linearly with loading. This supports the conclusion that for these samples, the magnetic response is directly proportional to the ferrite volume fraction. However, the response for the 25 wt%

loading sample is about a factor of two larger than one would expect from this scaling hypothesis. This response may be related to the enhancement effect of the dielectric according to the idea suggested in the introduction. On the other hand, one data point is an insufficient basis for such a general conclusion. More work is clearly needed to resolve this effect. The loss data to be considered shortly will demonstrate that the lower the loading, the more severe the degradation in the cavity Q due to the sample even in the absence of magnetic interactions. Such excessive loading introduces further complications. The interpretation of the K/m results may be more involved than simple scaling.

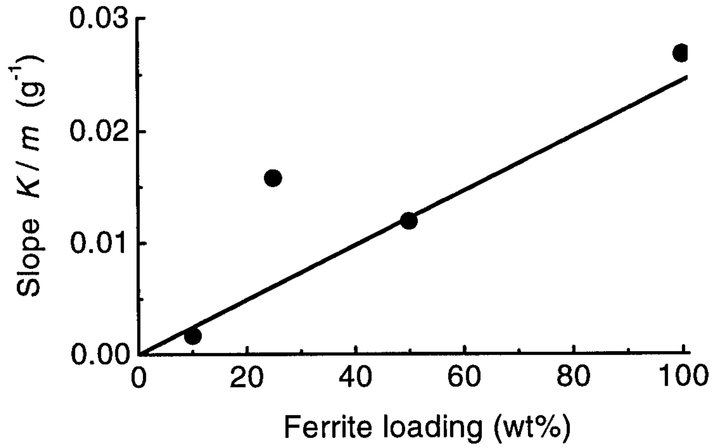


Fig. 6. Tail response slope parameter K/m as a function of the ferrite loading. The solid circles indicate the experimental points and the solid line shows the calculated theoretical response based on microwaveperturbation theory.

Turn now to the Q response. This preliminary work was not intended to provide a detailed study of the loss aspect of composite properties. As shown above, there are enough questions with regard to dispersion and scaling without including microwave loss in the analysis. As shown below, however, the loss results may have a direct bearing on the interpretation of the dispersion and scaling data. These data can also serve as a basis for future work on the intrinsic microwave losses in the ferrite phase of ferroelectric ferrite composites. As with the dispersion and scaling data, these results also raise important questions about the overall microwave response of ferroelectric composite materials.

Figure 7 shows plots of the inverse Q as a function of the absorption parameter X_Q for the 10 wt%, 25 wt%, 50 wt%, and 100 wt% loadings. These data are shown in raw form, without subtraction of the high field $1/Q_N$ term and without normalization to sample mass. The X_Q parameter was evaluated from the frequency vs. field data and the same ferrite parameters as indicated above. As expected from the response equations given in the previous section, $1/Q$ is a linear function of the absorption tail parameter X_Q for all the samples. On the other hand, the base nonmagnetic microwave loss which corresponds to the extrapolated $1/Q$ value at $X_Q = 0$ is extremely large. The 100 wt% ferrite loading sample with no ferroelectric has an inverse Q value of about 5×10^{-5}. This corresponds to a cavity Q of 20,000, which is close to the empty cavity Q. The pure ferrite sample, therefore, produces almost no change in the cavity Q at high field and well away from the FMR region. As the amount of the ferroelectric is increased, however, this high field Q-value drops substantially. For the 10 wt% loading, and the most ferroelectric content, this extrapolated Q drops by a factor of five or more. Note that this extrapolated Q to $X_Q = 0$ corresponds to the Q_N parameter in Eq. (2). This severe loading effect shows that the ferroelectric medium, at least for these samples, changes the cavity

response significantly. Clear conclusions about scaling for the microwave magnetic response, as considered above, will require samples without such losses.

In spite of these problems, the linear response and the analysis of the previous section may be used to obtain effective linewidths. Figure 8 shows the same data as in Fig. 7, except that the vertical axis now displays $(1/Q - 1/Q_N)/K$. The K parameter corresponds to the slope of the f vs. X_F plot for the sample. In accord with the analysis of the previous section, the slope of the plot for a given sample should correspond to the effective linewidth. The data show that these slopes for the 100 wt% and 50 wt% loadings are small while the 10 wt% and 25 wt% samples show large slopes. From smallest to largest, the slopes and effective linewidths from the data in Fig. 8 are 26 Oe, 43 Oe, 366 Oe, and 483 Oe, respectively.

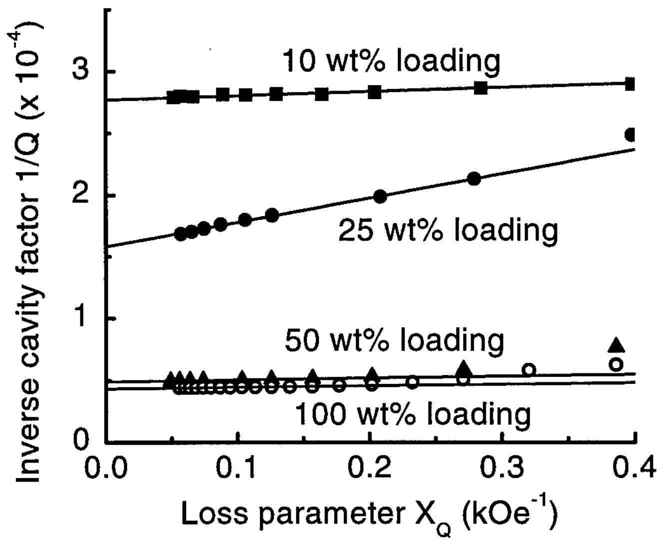


Fig. 7. Inverse Q vs X_Q tail response for the four loadings as indicated.

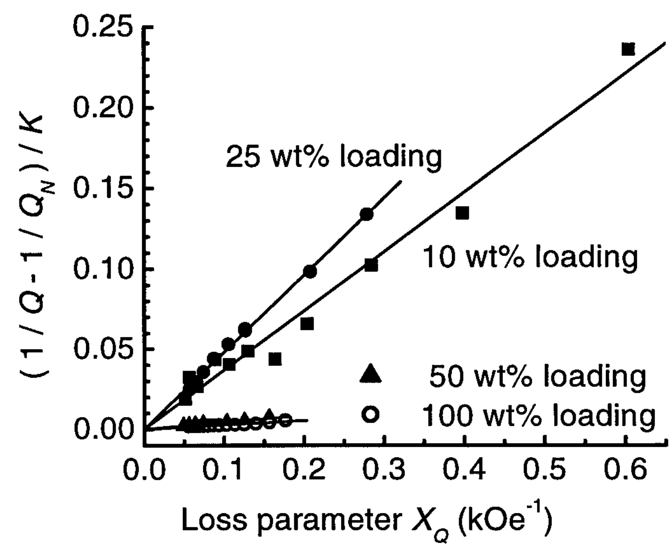


Fig. 8. Reduced data from the tail analysis for the absorptive part of the microwave susceptibility vs. field for the four loadings as indicated.

The ΔH_{eff} value of 26 Oe for the 100 wt% loading is consistent with the large 154 Oe linewidth from the Trans-Tech specifications. One generally expects the high field effective linewidth to be substantially smaller than the actual FMR linewidth. A value of 20-30 Oe is reasonable for the fired powder with some residual porosity (Patton, 1972, 1973). The somewhat larger but still respectable, value of 43 Oe for ΔH_{eff} in the 50 wt% loading material indicates that at this 50-50 ferrite-ferroelectric level, the basic ferrite component is still maintained intact. These relatively low effective linewidths are also somewhat surprising, in view of the highly distorted and broad FMR absorption curves in Fig. 4. The physical origin of these effective linewidths will be an important focus for a full program on ferroelectric ferrite composites. A first step, of course, will be to obtain homogeneous samples with clean and as narrow as possible FMR response curves.

The two samples with the lowest ferrite loadings at 10 wt% and 25 wt% show anomalously large effective linewidths. The origin of these large values is unknown. It is important to keep in mind that these samples have excessive electrical loss, as evidenced by the extremely large effect these samples have on the cavity Q at high field. These samples also have very broad and extremely distorted FMR absorption curves as shown in Fig 4. These problems will have to be resolved before one can address the problem of loss and effective linewidth in a quantitative way.

Table IV summarizes the effective linewidth results and provides comparisons with the FMR linewidth data in Fig. 4. The first row shows properties for the TT2-111 ferrite from Trans-Tech. The second row shows the realized linewidths for the 100 wt% loading sample which was fired in the same way as the composite samples. A comparison of these data show that the effective linewidth for the 100 wt% loading material is reasonably close to that expected for the ferrite, but that the actual FMR linewidth is much broader. These data alone demonstrate one clear problem for future work in terms of an optimization of the ceramic processing parameters for the ferrite. Up to now, the main work and progress at Paratek has been on optimization for the ferroelectric phase. It is not unreasonable to find that this processing is not optimum for the ferrite. Further work is clearly needed in this area. It is anticipated that small changes in the processing parameters will yield narrow FMR linewidths, reduced dielectric loading for the ferroelectric phase, and an even lower effective linewidth. Such materials will provide a sound series of composites for the determination of scaling, susceptibility enhancement, tunability, impedance matching, and other properties important for device implementation.

Table IV. Summary of 10 GHz linewidth results.

Loading wt% ferrite	High field effective linewidth ΔH_{eff} (Oe)	Ferromagnetic resonance linewidth ΔH (Oe)	Ferromagnetic resonance line shape
TT2-111 ferrite(1)	10-20(2)	150 - 200(3)	Lorentzian and symmetric
100	27	1200	Lorentzian and symmetric
50	43	1600	non-Lorentzian and symmetric
25	483	1400	Broad, shifted up in field, and distorted (steep on high field side)
10	366	1200	Broad, shifted down in field, and distorted (steep on low field side)

(1) Supplied by Trans-Tech.

(2) Estimated from effective linewidth results for polycrystalline large grain yttrium iron garnet materials (Patton, 1974).

(3) Range expected from Trans-Tech specifications.

II. SUMMARY AND CONCLUSION

In summary, the ferrite ferroelectric composites have the following properties:

1. The dielectric constant follows the basic scaling expected for an effective medium. This suggests that the composite can be used for impedance matching.

2. The dielectric loss tangent seems to increase for the composites, relative to either the pure ferroelectric or the pure ferrite. This indicated that further work on processing parameters is needed.

3. The static magnetization appears to scale with the wt% loading of the ferrite. This indicates that the ferrite enters the composite more or less structurally intact and the magnetization of the base ferrite component is preserved.

4. The saturation field appears to decrease as the wt% ferrite loading decreases. This is inconsistent with a simple model non-interacting spherical particle model for the ferrite phase. Interactions between ferrite particles would lead to a saturation field which (1) would be $4\pi M_s/3$ in the limit of 100 wt% loading and in the limit of small loadings, and (2) show a minimum at some intermediate loading value.

5. The ferromagnetic resonance linewidths are generally broad and distorted, and are about a factor of 8 larger than the specification linewidths for the base ferrite. This indicates (1) impurity and inhomogeneous broadening contributions to the microwave losses, and (2) complicated microstructure for the ferrite phase of the composite. The large linewidths may also be related to the high dielectric loss.

6. The high field dispersion tail analysis indicates a reasonable uniform mode ferromagnetic resonance response. The data indicate that the response scales with the ferrite loading, except for the 25 wt% loading. This critical data point holds the key to the possible ferroelectric enhancement in the microwave magnetic response. Further work on this aspect of the response is needed.

7. The high field effective linewidth analysis yields reasonable values for the 50 wt% and 100 wt% loadings and very large values for the 10 wt% and 25 wt% loadings. Here too, these data indicate the need to optimize processing parameters, reduce dielectric loss, and refine the microstructure of the ferrite phase.

In conclusion, this STIR program has established a starting point for a full scale research effort on ferroelectric ferrite composite materials for possible microwave and millimeter wave applications. The results so far are encouraging that suitable low loss materials can be produced which are voltage tunable and magnetic field tunable, and which are capable of providing good impedance matching and small size. A proposal for a full scale research effort to accomplish these goals is in preparation.

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